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A CRITICAL ANALYSIS OF GRAIN-SIZE AND YIELD-STRENGTH DEPENDENCE--ETC(U)
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Near-threshold fatigue-crack growth behavior has been analyzed for a broad selection of steels surveyed from the literature. It is clear first of all that apparent values of the threshold stress-intensity factor (ΔK_{th}) can vary widely, roughly an order of magnitude. Though in many instances actual ΔK_{th} values are difficult to define rigorously, a pronounced transition point or "knee" is apparent in the near-threshold region of the conventional logarithmic plot of fatigue crack growth rate (da/dN) as a

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function of stress-intensity range (ΔK). Though the values of ΔK associated with these transition points (ΔK_T) for an individual steel may tend to exhibit a functional dependence on yield strength (σ_{ys}) or grain size ($\bar{\ell}$) -- as is the case, for example, with a low-carbon ferritic steel -- it is unmistakably clear that for the gamut of steels examined (15 cases), the transition points do not order on the basis of either σ_{ys} or $\bar{\ell}$ alone. Rather, values of ΔK_T for the gamut of steels order on the basis of a synergetic interaction of σ_{ys} and $\bar{\ell}$, according to the equation, $\Delta K_T = 5.5\ \sigma_{ys}\sqrt{\bar{\ell}}$. This relationship was derived in the cyclic plastic zone model of fatigue crack growth established in our prior work with titanium alloys. In further agreement with this model, ΔK_T has been identified for these steels as the point at which the cyclic plastic zone attains the mean grain size. The significance and implications of these findings appears far-reaching, as the steels surveyed include those of both high and low strength, a wide range of effective grain sizes (mean free path in the case of high-strength steel), and a host of microstructural types (ferritic, martensitic, pearlitic, bainitic, austenitic).

CONTENTS

"A CRITICAL ANALYSIS OF GRAIN-SIZE AND YIELD-STRENGTH DEPENDENCE OF NEAR-THRESHOLD FATIGUE-CRACK GROWTH IN STEELS"

INTRODUCTION

Though a number of investigators have examined the influence of microstructural variables on near-threshold fatigue-crack growth rates in steels, a comprehensive understanding of the dependence of near-threshold growth rates on grain size, yield strength and microstructural morphology in steels has yet to emerge -- as noted in an excellent review by Ritchie [1]. Recently, however, from our own extensive studies with α/β titanium alloys, the basis for microstructural dependence of widely different fatigue crack growth rates was established for titanium alloys [2-6]. Inasmuch as the micromechanistic model from that work does not depend uniquely on alloy family, it is of great interest to explore its *applicability to steels* -- especially since it predicts quantitatively the influence of yield strength and grain size in the near-threshold region for steels. Thus, the purpose of this paper is to critically analyze the near-threshold fatigue crack growth behavior, as reported in the literature, for steels of widely different strength level, grain size and microstructural morphology -- in the search for a systematic ordering of near-threshold fatigue crack growth rates that pertains to the whole gamut of steels.

SURVEY AND ANALYSIS

A summary of the near-threshold fatigue-crack growth behavior surveyed from the literature is presented in Fig. 1, together with symbols which identify the steel and respective investigator(s) in Table I [7-15]. For each of the 15 materials represented in this logarithmic plot of fatigue crack growth rate (da/dN) vs. stress-intensity range (ΔK), the growth-rate data exhibit a bilinear form with a transition point or "knee" as illustrated in Fig. 1. The level of stress-intensity range associated with the transition

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point is designated as ΔK_T. Since the observed value of ΔK_T for each material in the figure is unique, it serves in Table I to distinguish the yield strength (σ_{ys}) and grain size ($\bar{l}$) of that material in those instances where a given symbol pertains to multiple growth-rate curves. It is important to point out that virtually all of the growth-rate data represented in this figure were generated at roughly the same value of stress ratio, viz. $0.0 \le R \le 0.1$.[1]

As shown in Fig. 1, apparent values of the threshold stress-intensity range (ΔK_{th}) can vary widely among the different steels, by roughly as much as an order of magnitude.[2] Though in many instances actual ΔK_{th} values are difficult to define rigorously, Fig. 1 indicates a spectrum of values from about 3 to 18 $MPa{\cdot}m^{1/2}$. Note, however, that in the case of each material, the transition point ΔK_T closely approximates ΔK_{th}, since the slope of the hypotransitional branch of the growth-rate curve is so steep -- i.e. the exponent in the growth rate law [18],

$$da/dN = C(\Delta K)^m \tag{1}$$

is very large. Inasmuch as the transition points ΔK_T are found to order systematically for the 15 materials in Fig. 1, on the basis of a synergetic interaction of the yield strength and grain size -- to be elucidated in this paper, this point is of potentially prime engineering significance with regard to the estimation of thresholds ΔK_{th}.

The broad spectrum of ΔK_T values represented in Fig. 1 derives from steels that vary widely in terms of composition, microstructural morphology, yield strength and grain size, as indicated in Table I. Microstructural types range, for example, from basically martensitic 4Cr - 0.35C steel to primarily ferritic 1005, 1007 and 1020 steels, pearlitic 1055 steel or austenitic 304 stainless, etc. Both high and low strength steels are represented, with yield strengths ranging from 192 MPa $\le \sigma_{ys} \le$ 1324 MPa.

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1 The data of Taira, Tanaka and Hoshina [15] are treated as though R = 0, as the authors originally published.

2 Earlier surveys of ΔK_{th} [16, 17] which were based upon more limited data did not reveal the broad range of near-threshold behavior that is now evident.

Effective grain sizes (transformed grain size or mean free path in the case of high-strength steel) range from 0.38 $\mu m \leq \bar{\ell} \leq 76$ μm.[3]

In prior work with low-strength steels, it has been reported that fatigue crack growth rates decrease with increased grain size and (or) that the threshold ΔK_{th} itself increases with increased grain size [7, 12, 15, 21-24]. Figure 2 illustrates behavior found typically for a particular steel: Though the said effects appear operative as the grain size is increased through heat treatment of the given plate of steel, it is also clear that the yield strength (σ_{ys}) has been simultaneously decreased. Thus the isolation of any grain-size effect is obscured, as pointed out by Ritchie [1] and Benson [7]. Others have also noted a decrease in ΔK_{th} with an increase in σ_{ys} [12, 24-26]. Though the data in Fig. 2 will be further considered in the latter stages of this paper, it is worthwhile to note at this point the degree to which the data support the bilinear form of the growth-rate curves shown here and in Fig. 1.

In contrast to Fig. 2, if behavior for the broad spectrum of steels described in Fig. 1 and Table 1 is examined, there is no clearly discernible influence of σ_{ys} and $\bar{\ell}$ on the

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3 For the basically ferritic steels or the 304 stainless, $\bar{\ell}$ is the ferrite or austenite grain size, respectively. For the 1055 steel, $\bar{\ell}$ is the pearlite colony size, since all α-phase lamellae in a colony exhibit the same variant of the Kurdjumov-Sachs transformation relationship -- so that, accordingly, slip bands or slip-band cracks can transmit across α-lamellae with carbide platelets cracking very readily under tensile load [10, 19, 20] as though the colony were a pseudo single crystal. In the case of the dual-phase, ferritic-martensitic 1018 steel, which is nearly 65% ferrite, $\bar{\ell}$ is taken as the ferrite grain size, consistent with Suzuki and McEvily's observation that crack growth preferentially followed a path through the ferrite [14]. For the martensitic 4Cr - 0.35C steel, $\bar{\ell}$ would be the size of the packet of martensite laths which have a common variant according to the Kurdjumov-Sachs or the Nishiyama-Wasserman transformation relations -- were there no austenite remaining at the lath boundaries as an effective barrier to the transmission of slip bands between adjacent laths [9]. In those cases where virtually continuous films of retained austenite line the lath boundaries (viz. for austenitizing temperatures of 1000, 1100 and 1200°C), values of $\bar{\ell}$ are taken as the lath widths reported by Carlson, Narasimha Rao and Thomas, viz. $\bar{\ell}$ = 0.47, 0.41 and 0.38 μm, respectively. By contrast, for the material quenched from the lowest austenitizing temperature (870°C), the austenite film is highly discontinuous; in this instance, the appropriately modulated value of mean free path is estimated (from quantitative metallographic methods by the present authors) from the thin-film transmission electron micrograph in Ref. [9] to be $\bar{\ell}$ = 0.83 μm.

ordering of the growth-rate curves, when each variable is considered separately. This point is illustrated in Fig. 3, where a random sampling of data from Fig. 1 and Table 1 is represented. As the growth-rate curves (in terms of ΔK_{th} or ΔK_T) are traversed from left to right, values of $\bar{\ell}$ exhibit no consistent trend, nor do those of σ_{ys}. When the functional dependence of ΔK_T on σ_{ys} and $\bar{\ell}$ is examined for all 15 of the materials of Fig. 1 and Table 1, as shown in Figs. 4 and 5 respectively, again the data patterns of ΔK_T vs. σ_{ys} and $\bar{\ell}$ appear highly random. For the sake of completeness, data from our own work with titanium alloys [4, 27] have been included in these and subsequent plots, denoted by the symbol "X". All other data symbols in these figures are identified in Table 1.

If ΔK_T does not order on the basis of either σ_{ys} or $\bar{\ell}$, when the full gamut of steels is considered, then what is the system to the ordering of the growth-rate curves *in the near-threshold region? Happily, there appears to be a straightforward answer to* this question: In accord with the cyclic plastic zone model for the ordering of growth-rate curves, as developed from work with titanium alloys [2-6], observed values of ΔK_T for the whole spectrum of steels examined in this study are in remarkable agreement with the quantitative predictions of that model. In further agreement with the model, values of $\bar{\ell}$ for all these steels, independent of microstructural morphology or composition, are found equal to respective values of the cyclic plastic zone size at the transition point, ΔK_T.

To begin with: In the work with titanium alloys, a bilinear growth-rate behavior was observed as illustrated in Fig. 6, with two distinct branches that independently obey the power law of Eq. (1) and which join together at the transition point (T). In the hypotransitional region where the cyclic (or "reversed") plastic zone size (sketched for plane strain conditions) is less than the grain size, $r_y^c < \bar{\ell}$, a microstructurally sensitive (or "structure-sensitive") mode of crack growth occurs that involves crystallographic bifurcation in grains adjacent to the Mode I crack plane. This bifurcation causes a reduction in the effective ΔK and consequently da/dN, and thus the appearance of the transition itself. By contrast, in the *hypertransitional region* where $r_y^c > \bar{\ell}$, the grains within the larger r_y^c must necessarily deform as a continuum, which results in a microstructurally insensitive, nonbifurcated mode of crack growth [4, 5]. It should be noted that bifurcation has been observed in the microstructurally sensitive region for a wide range of microstructures [28, 29]. Moreover, it is appropriate to mention that Wanhill and Döker [30] have observed a difference in the

dislocation substructures associated with the structure-sensitive and structure-insensitive modes of crack growth. Quantitatively, a shift in the fatigue crack growth rate curve can be predicted, since the transition point (T) in Fig. 6 is the point at which the reversed plastic zone size [31-33],

$$r_y^{\,c} = 0.033\,(\Delta K/\sigma_{ys})^2 \tag{2}$$

attains the mean grain size [2-6, 27-29]. Thus at the transition point, where $r_y^{\,c} = \bar{\ell}$, it follows that

$$\Delta K_T = 5.5\,\sigma_{ys}\sqrt{\bar{\ell}} \tag{3}$$

This shift in the da/dN data plot, as sketched in Fig. 7, is well documented for titanium alloys [2-6, 27-29].

To examine the applicability of this model to the near-threshold fatigue crack growth behavior of the whole gamut of steels described by Fig. 1 and Table 1, values of $\bar{\ell}$ are compared in Fig. 8 to respective values of the cyclic plastic zone size from Eq. (2), computed at the transition point, ΔK_T. The agreement appears to be excellent. In further accord with Fig. 6, a number of investigators have reported a structure-senstive mode of crack growth when the cyclic plastic zone size was less than the grain size, which transitioned to a structure-insensitive mode of crack growth when the cyclic plastic zone exceeded the grain size; for example, cf. Aita and Weertman [19, 20], Suzuki and McEvily [14], and Cooke, Irving, Booth and Beevers [34]. Finally, to examine whether the wide spread in near-threshold growth-rate curves in Fig. 1 is simply a consequence of the "omega" shift predicted in Fig. 7 via Eq. (3), values of ΔK_T observed for the whole gamut of steels in Fig. 1 are compared in Fig. 9 to values of ΔK_T computed from Eq. (3). The level of agreement is considered remarkable -- particularly in view of uncertainties involved in the measurements made by different investigators.[4] Thus the broad spectrum of near-threshold growth-rate curves for the

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[4] For example, inasmuch as there is as yet no ASTM standard for the relatively difficult measurement of growth rates in the near-threshold region, the data represented in Fig. 1 were obtained by a multitude of different procedures. Moreover, as pointed out in a recent paper by Vosikovsky, Trudeau and Rivard [35], residual stresses can enormously affect determinations of apparent near-threshold growth rate behavior.

gamut of 15 steels in Fig. 1 clearly orders in accord with the predictions of Fig. 7 and Eq. (3).

DISCUSSION

Evidence has been presented for a systematic ordering of near-threshold growth rates for steels, based on the synergetic interaction of yield strength (σ_{ys}) and grain size ($\bar{\ell}$) expressed by Eq. (3). Inasmuch as this finding pertains to the whole gamut of steels, regardless of strength level, microstructural morphology or composition, it would appear to be extraordinarily significant. In particular, from an engineering standpoint, it is suggested that estimation of the threshold for fatigue crack growth ΔK_{th} ought to be considered via Eq. (3) —which requires knowledge of only σ_{ys} and $\bar{\ell}$, to obviate the need for direct measurements of ΔK_{th}, which though elegant, are very time-consuming and expensive.

Though the near-threshold growth-rate curves for the whole gamut of steels do not order on the basis of grain size alone, it is pertinent to readdress the case of an individual steel, heat-treated to generate different levels of $\bar{\ell}$ (and σ_{ys}) as illustrated in Fig. 2. Since the Hall-Petch relation provides the tradeoff between σ_{ys} and $\bar{\ell}$ in steels [36, 37],

$$\sigma_{ys} = \sigma_o + k(\bar{\ell})^{-\frac{1}{2}} \tag{4}$$

where σ_o and k are material constants, then for an individual steel it follows from Eq. (3) that

$$\Delta K_T = 5.5\,(\sigma_o\sqrt{\bar{\ell}} + k) \tag{5}$$

or,

$$\Delta K_T \;\alpha\, \sqrt{\bar{\ell}} \tag{6}$$

Thus it is possible to understand micromechanistically the "grain-size" effects reported by Taira, Tanaka and Hoshina [15] and Masounave and Bailon [12], viz.

$$\Delta K_{th}\, \alpha \sqrt{\bar{\ell}} \tag{7}$$

It is of further interest to note that in the case of titanium alloys, the Hall-Petch relation, Eq. (4) is inoperative — i.e. the dependence of σ_{ys} on $\bar{\ell}$ is very weak, at best [3, 38]. Consequently, the $\sqrt{\bar{\ell}}$ term dominates the right-hand side of Eq. (3), so that a grain-size effect is observed for titanium alloys [2-4, 21, 38, 39]. In that case, the

inverse dependence of fatigue crack growth rates upon grain size is directly relatable to the microstructurally sensitive mode of crack growth --since larger bifurcated cracks (thought to be slip-band cracks [5]) occur as the grain size is increased, thus dispersing the strain-field energy of the macroscopic crack over increased volumes of material in the crack-tip region --to further reduce the effective ΔK and consequently, da/dN.[5]

As shown by Hertzberg and Mills [41] for a wide range of alloy families, slip-band cracking or "decohesion" is characteristic of near-threshold growth-rate behavior, or the microstructurally sensitive mode of fatigue crack growth. For steels, Taira, Tanaka and Nakai [42] have developed a blocked slip band model, with particular reference to the results of Ref. [15], in which the ferrite grain boundaries served as the blocking obstacles.[6] If the structure-sensitive mode of crack growth can occur only so long as the cyclic plastic zone size is less than the maximum dimension to which a slip-band (or slip-band crack) can develop unimpeded by an (insurmountable) obstacle, then $\bar{\ell}$ must in the general sense represent the mean free path between such obstacles. For α/β titanium alloys and low-strength steels, $\bar{\ell}$ is thus the effective grain size -- inasmuch as the grain boundaries are the controlling obstacles. In the case of the high-strength steels of Carlson et al [8, 9], as noted above in Footnote 3, the appearance of retained austenite in the martensite lath boundaries truncates the mean free path for slip band transmission, which would otherwise be the lath packet dimension.

If application of the model represented by Eq. (3) and Fig. 7 were to be extended to the near-threshold, transitional growth-rate behavior observed in precipitation-hardened alloys such as those in the aluminum family [44-47] or others [48], thin-foil transmission electron microscopy might well be required to determine the mean free path $\bar{\ell}$ between obstacles that effectively obstruct slip-band transmission. Such

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5 In recent work by J. P. Lucas and W. W. Gerberich with an HSLA steel, the Hall-Petch relation was similarly found to be inoperative for $\bar{\ell}$ in excess of 50 μm, with a large grain-size effect observed on ΔK_{th} [40]; moreover, a dislocation model for the ΔK_{th} behavior has been developed.

6 Recently Sadananda [43] has developed a dislocation model to explain the crystallographic, faceted mode of fatigue crack growth.

potential obstacles might include constituent or intermetallic particles, dispersoids or precipitates formed upon aging.[7]

In closing this discussion, it is appropriate to at least mention a further pair of issues concerning ΔK_T. On the one hand, in view of the work of Wilhem [50] some years ago, the question might be raised as to whether ΔK_T reflects a tensile to shear mode fracture transition. The answer appears to be a resounding no, since it appears that shear lip development was not involved in the case of any of the 15 materials in Fig. 1 and Table 1. Moreover, calculation of the limits for plane strain constraint indicates that levels of ΔK_T are nowhere near the calculated limits, except for possibly 1 or 2 of the 15 cases. On the other hand, the question of stress-ratio (R) influence on ΔK_T might be raised. From a micromechanics standpoint, it is not clear that R should have any influence on ΔK_T as formulated in Eq. (3). Rather, there is growing evidence that stress-ratio effects may be primarily environmental in their influence on the near-threshold growth-rate behavior: Witness, for example, the results for steels, aluminum and titanium alloys which indicate that ΔK_{th} (or ΔK_T) is insensitive to R in an inert environment, viz. vacuum conditions [1, 34, 45]. Moreover, it is relevant to note that ΔK_{th} values reported for vacuum over a wide range of R values are similar to ΔK_{th} *values observed near R = 0 for an air environment.* Thus the case can be made that environmental influence had negligible bearing on the results for the 15 steels analyzed in this paper.[8]

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7 Hertzberg has raised the question as to whether subgrain boundaries might act as the controlling obstacles in an extruded aluminum alloy [49].

8 The interested reader might wish to consult the work of Krafft [51] regarding stress-ratio effects and environmental modeling.

CONCLUSIONS

1. Resistance to fatigue crack growth in the near-threshold region varies widely among steels. When logarithmic plots of growth rate (da/dN) vs. stress-intensity range (ΔK) are analyzed in a bilinear form with a transition point or "knee" at ΔK_T, values of ΔK_T are observed to range from 3 to 18 $MPa{\cdot}m^{1/2}$ for the different steels. These values of ΔK_T appear to be good approximations to actual threshold values of ΔK below which cracks do not propagate (ΔK_{th}).

2. Values of ΔK_T for the gamut of steels examined do not order on the basis of either yield strength (σ_{ys}) or grain size ($\bar{\ell}$) alone. Rather, values of ΔK_T order on the basis of the *synergetic* parameter, $\sigma_{ys}\sqrt{\bar{\ell}}$.

3. Specifically, in accord with the cyclic plastic zone model developed in prior work with titanium alloys, observed values of ΔK_T are in remarkable agreement with predictions according to the equation, $\Delta K_T = 5.5\ \sigma_{ys}\sqrt{\bar{\ell}}$.

4. Applicability of these findings appears to be far-reaching, as the steels surveyed span a wide variety of yield strengths (192 to 1324 MPa), effective grain sizes (0.4 to 76 μm) and microstructural types (ferritic, martensitic, pearlitic, bainitic, austenitic).

5. A simple predictive equation (cf. Conclusion 3) is thus offered as a reasonable, but conservative approximation of the threshold, ΔK_{th} -- for any steel, from *knowledge of only* σ_{ys} and $\bar{\ell}$.

6. Though near-threshold growth-rate curves for the whole gamut of steels do not order on the basis of grain size alone, for the case of an individual steel, by contrast, ΔK_T does increase with $\sqrt{\bar{\ell}}$. This result is shown to be a direct consequence of the Hall-Petch relation, in combination with the equation for ΔK_T given in Conclusion 3.

7. The grain-size parameter in the cyclic plastic zone model, viz. $\bar{\ell}$, is recognized in the more general sense to be the mean free path between (insurmountable) obstacles to slip-band transmission.

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Table 1 — Identification of Materials

Symbol	Steel	Micro-Structure*	R	f (Hz)	$\bar{l}$ (μm)	σ_{YS} (MPa)	Observed ΔK_T (MPa · $m^{1/2}$)	Predicted ΔK_T (MPa · $m^{1/2}$)	Investigators
●	1005	F	0.1	100	24	411	10.3	11.07	Benson [7]
					70	368	12.8	16.93	
●	4 Cr — 0.35 C	M + A + C	0.05	50	0.83	1324	5.09	6.63	Carlson & Ritchie [8] (cf. Carlson, Narasimha Rao & Thomas [9])
					0.47	1324	3.82	4.99	
					0.41	1303	3.66	4.59	
					0.38	1324	3.57	4.49	
Δ	1055	P	0.05	85	27	399	14.4	11.42	Cooke & Beevers [10]
O	304 Stainless	A	0.0	0.07	76	273	17.8	13.09	James & Schwenk [11]
○	1007	F	0.1	50	19	219	9.2	5.25	Masounave & Bailon [12]
					40	192	9.4	6.68	
▽	2-1/4 Cr — 1 Mo	B + F	0.05	50	36	290	9.5	9.57	Ritchie et al. [13]
▷	1018	F + M	0.05	1-30	36	293	9.8	9.67	Suzuki & McEvily [14]
▲	1020	F + P	−1.0	30	7.8	366	5.47	5.62	Taira, Tanaka & Hoshina [15]
					20.5	275	6.47	6.85	
					55	194	7.95	7.91	

*A = Austenite, B = Bainite, C = Carbide (Fe_3C), F = Ferrite, M = Martensite, P = Pearlite

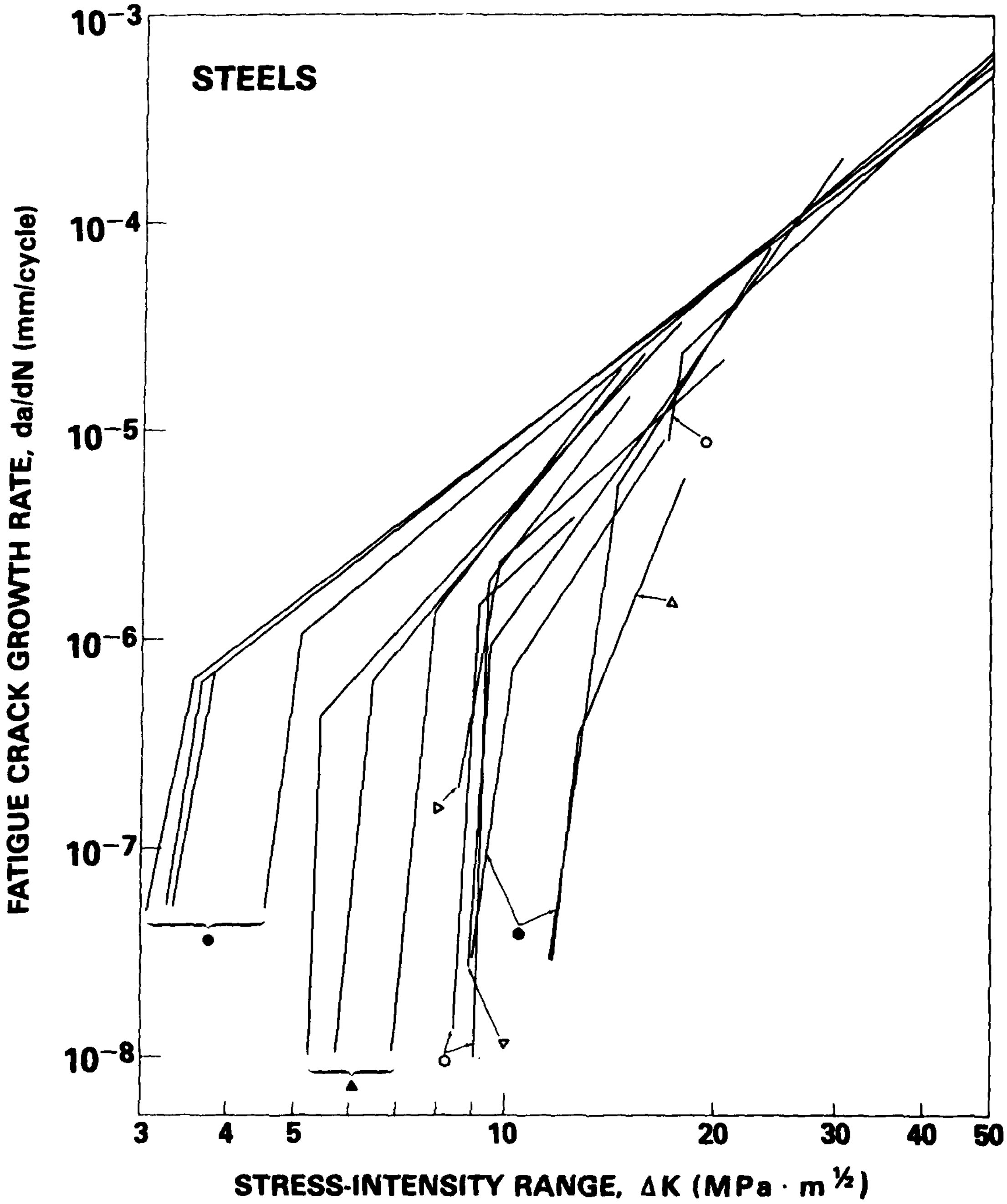

Fig. 1 Near-threshold fatigue-crack growth behavior of a broad selection of steels. Symbols refer to identification in Table I, cf. text for further details.

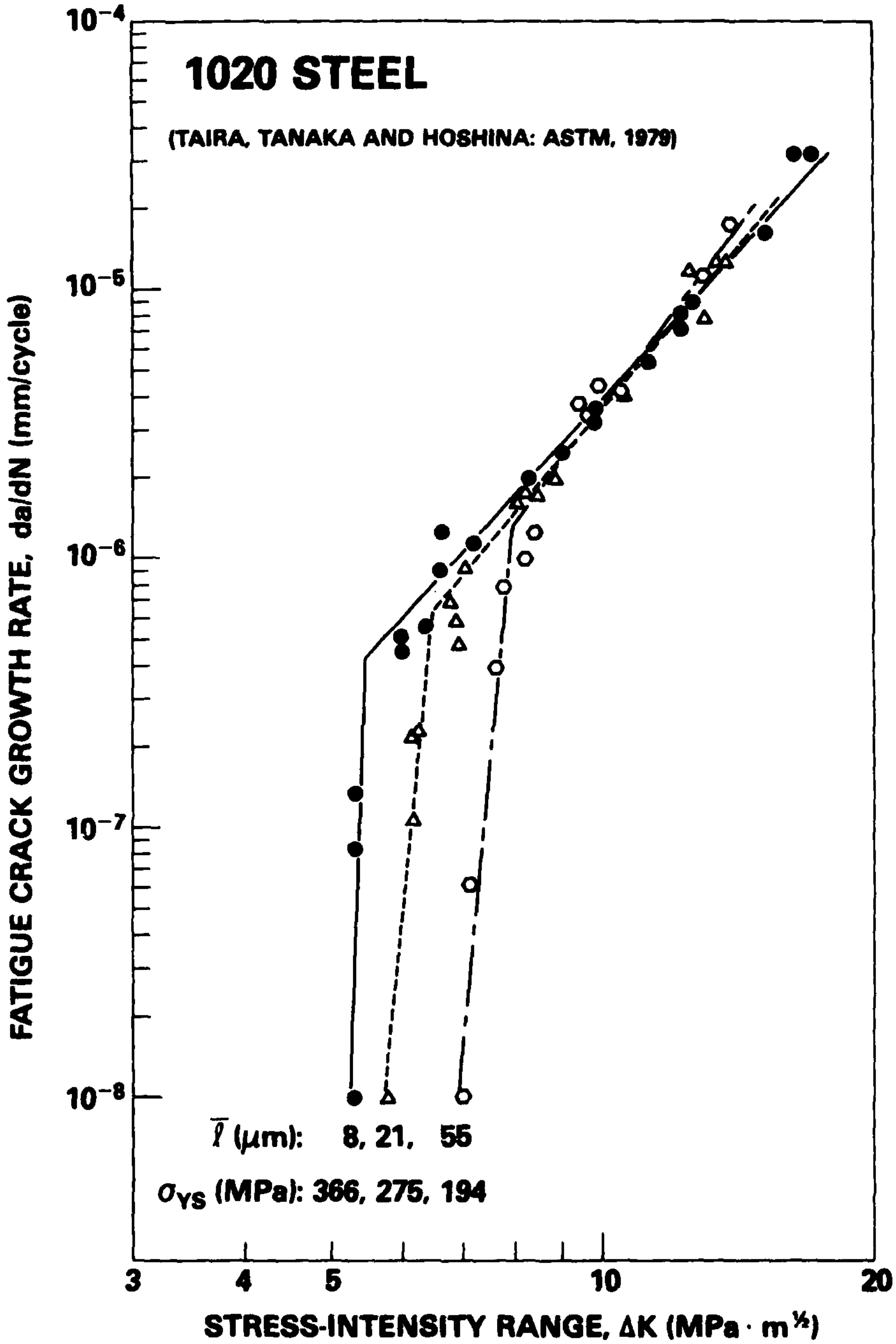

Fig. 2 Near-threshold fatigue-crack growth behavior of a 1020 steel heat-treated to the indicated levels of grain size ($\bar{l}$) and yield strength (σ_{ys}), after Ref. [15]. (Adapted from Fig. 11 (page 151), "Grain Size Effect on Crack Nucleation and Growth in Long-Life Fatigue of Low-Carbon Steel," *Fatigue Mechanisms,* ASTM STP 675, 1979.)

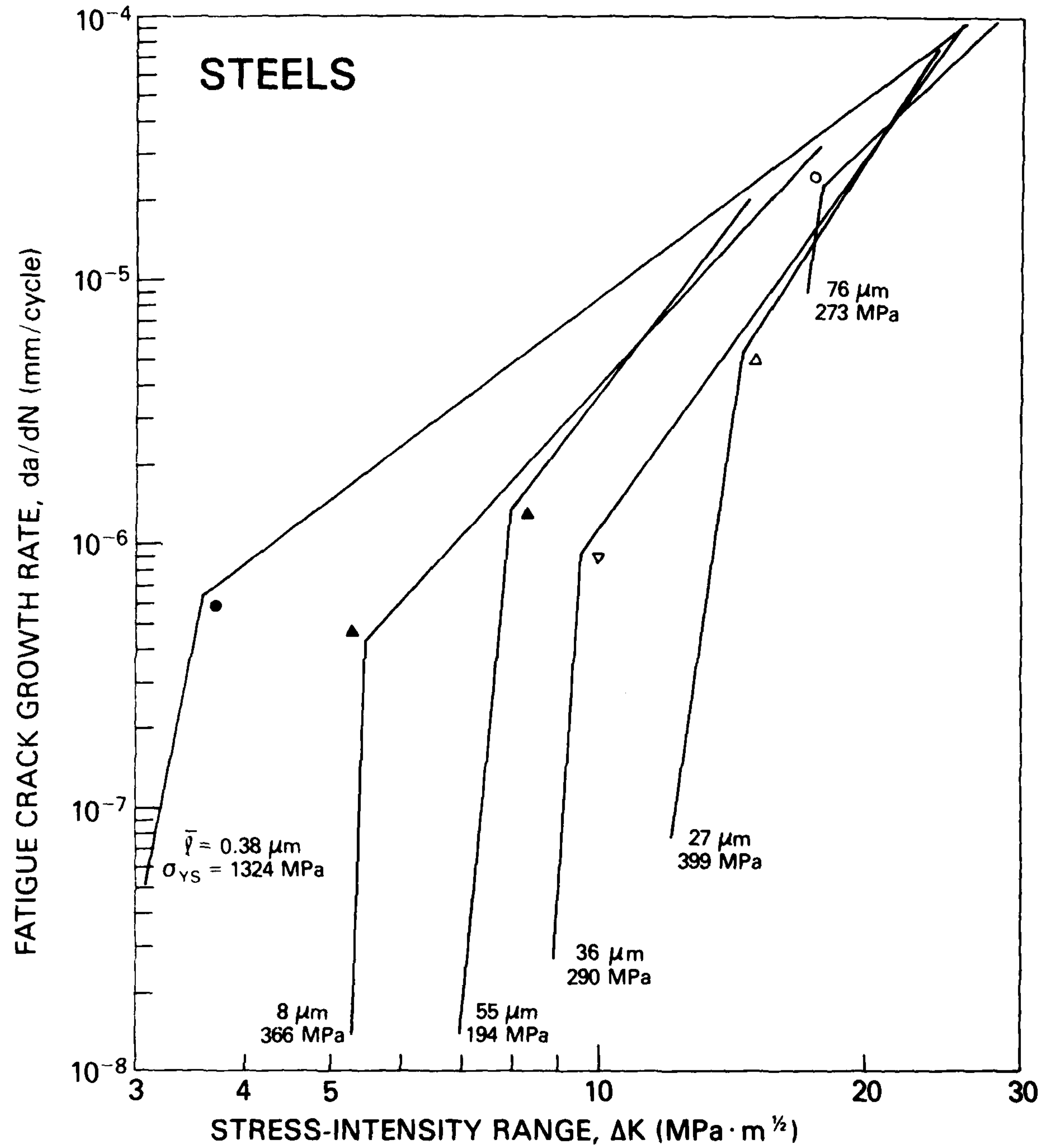

Fig. 3 As illustrated with selections from Fig. 1 (and Table 1), near-threshold growth-rate behavior for the whole gamut of steels fails to order on the basis of either grain size ($\bar{l}$) or yield strength (σ_{ys}) alone.

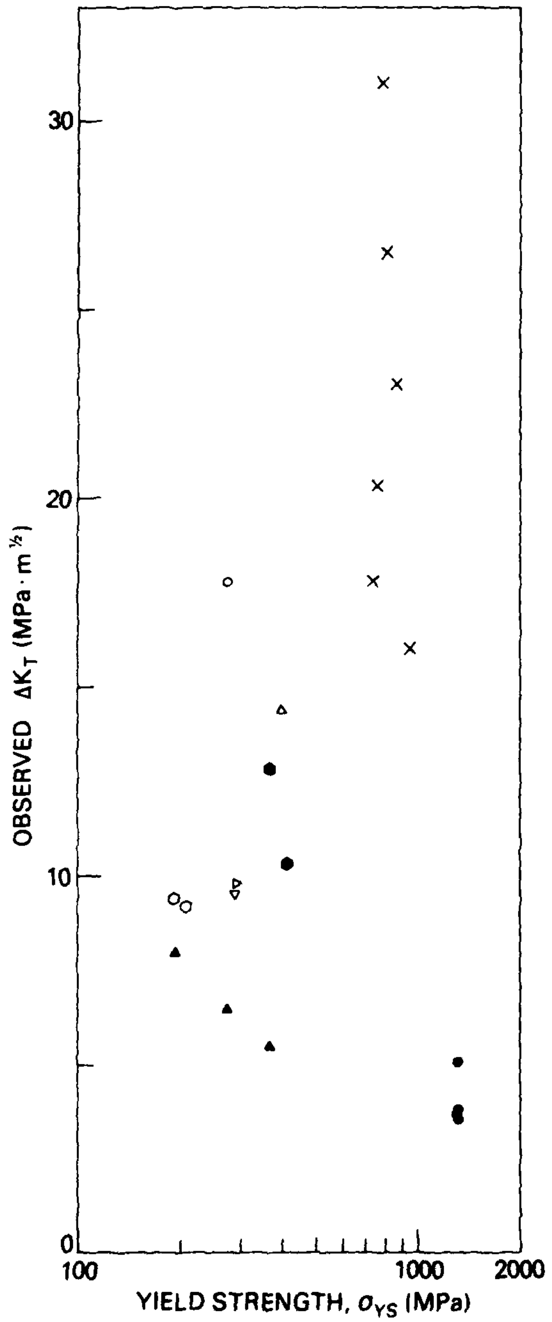

Fig. 4 Values of stress-intensity range observed at transition points or "knees" in the growth-rate data trend lines (ΔK_T) of Fig. 1 vs. yield strength (σ_{YS}). Symbols refer to Table I; additional symbol "X" refers to data from refs. [4, 27].

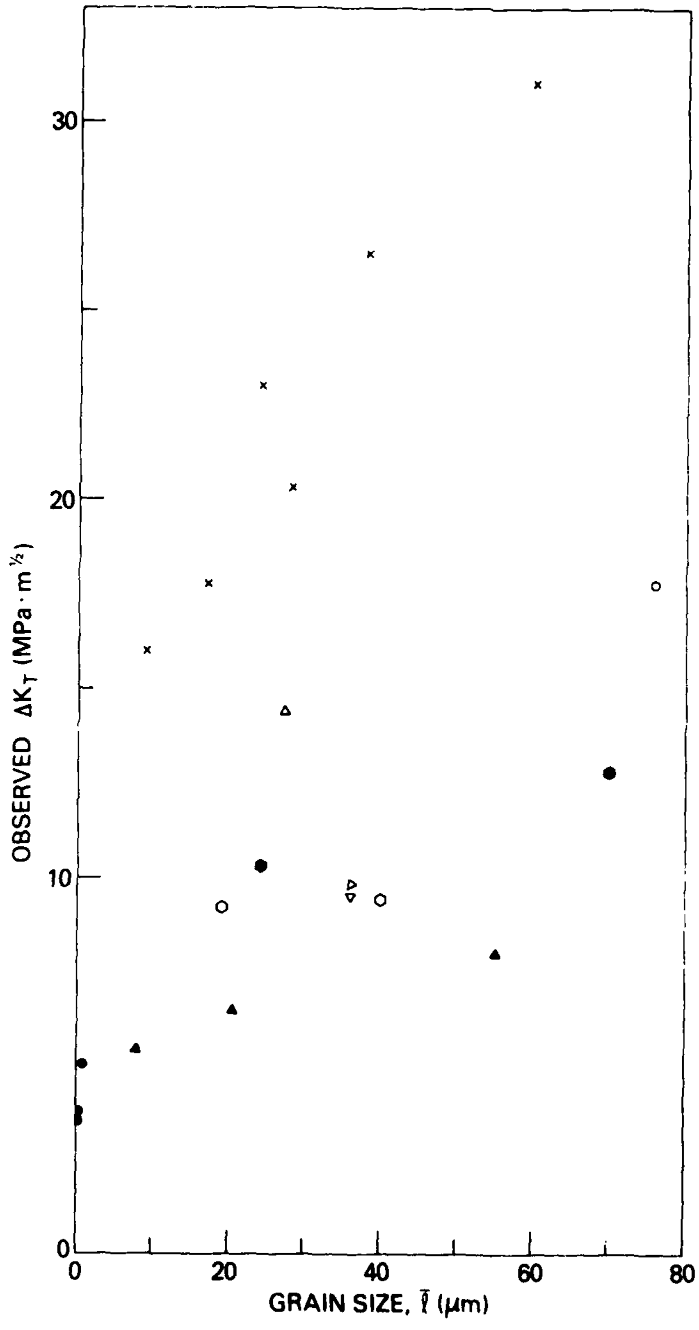

Fig. 5 Observed values of transitional stress-intensity range (ΔK_T) vs. effective grain size ($\bar{l}$).

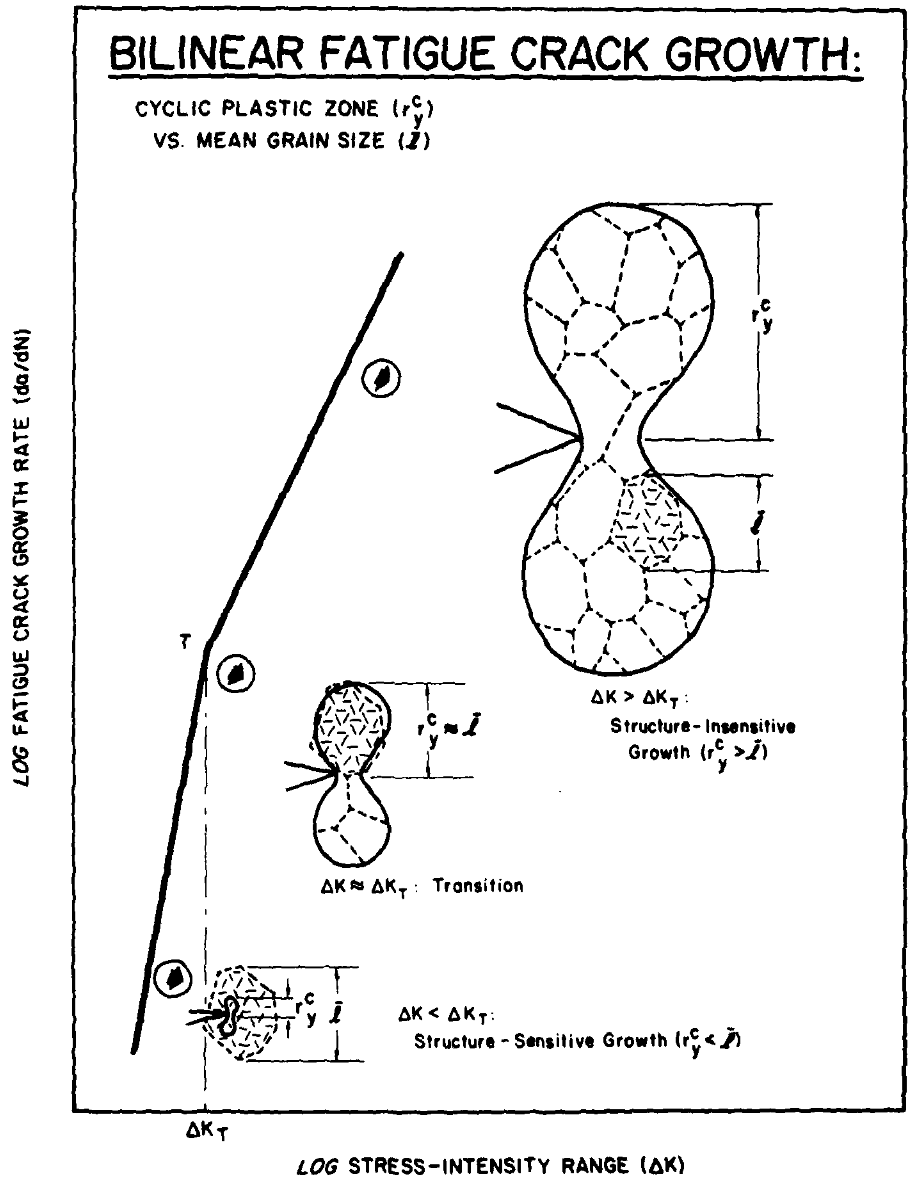

Fig. 6 Influence of cyclic (reversed) plastic zone size, relative to grain size, upon development of bilinear fatigue crack growth behavior. Note transition from structure-sensitive mode of crack growth in lower branch to structure-insensitive mode in upper branch.

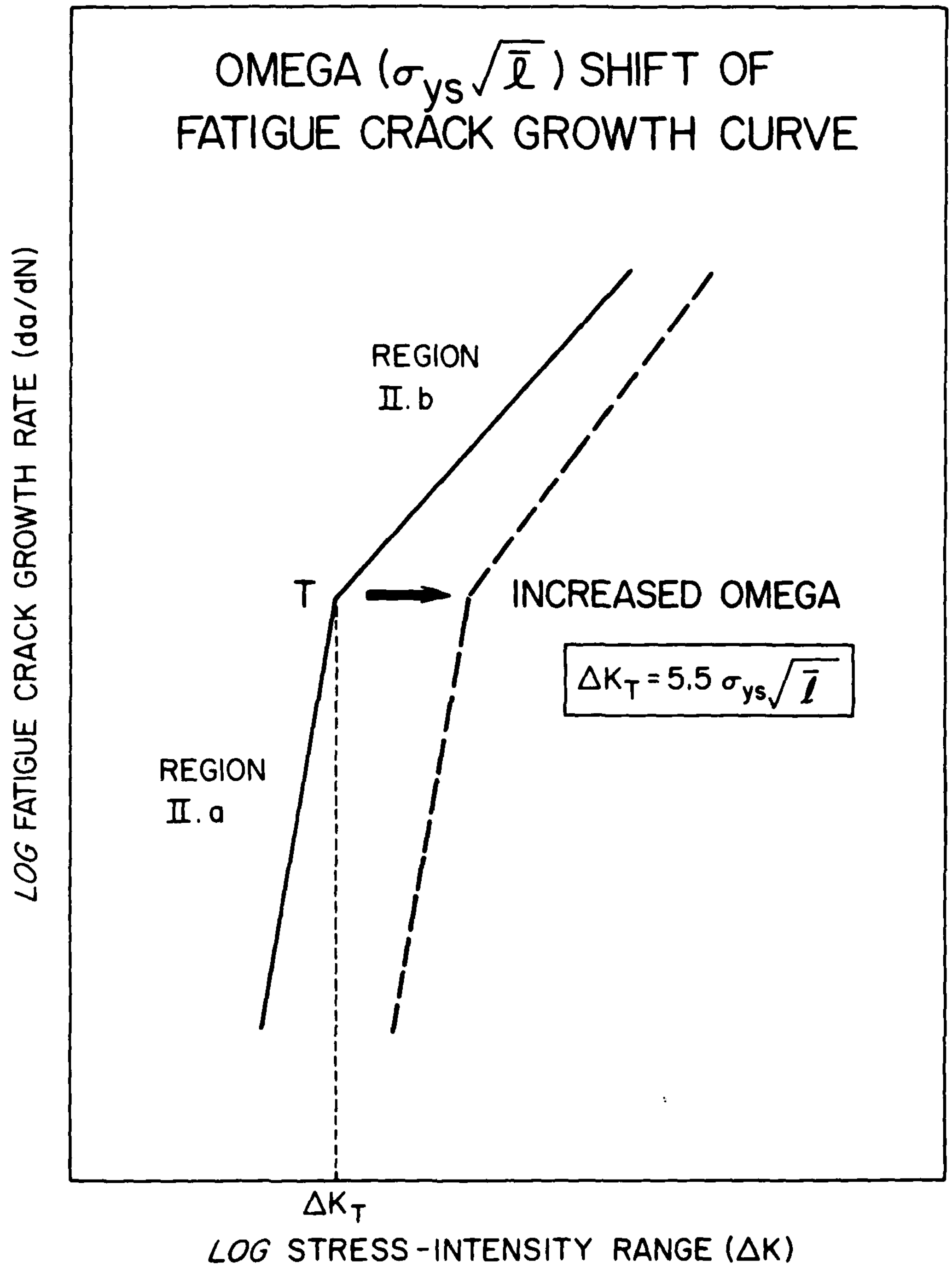

Fig. 7 From Fig. 6 and Eq. (3), shift in the fatigue-crack growth rate curve is predicted quantitatively from the synergetic interaction of yield strength (σ_{ys}) and grain size ($\bar{\ell}$).

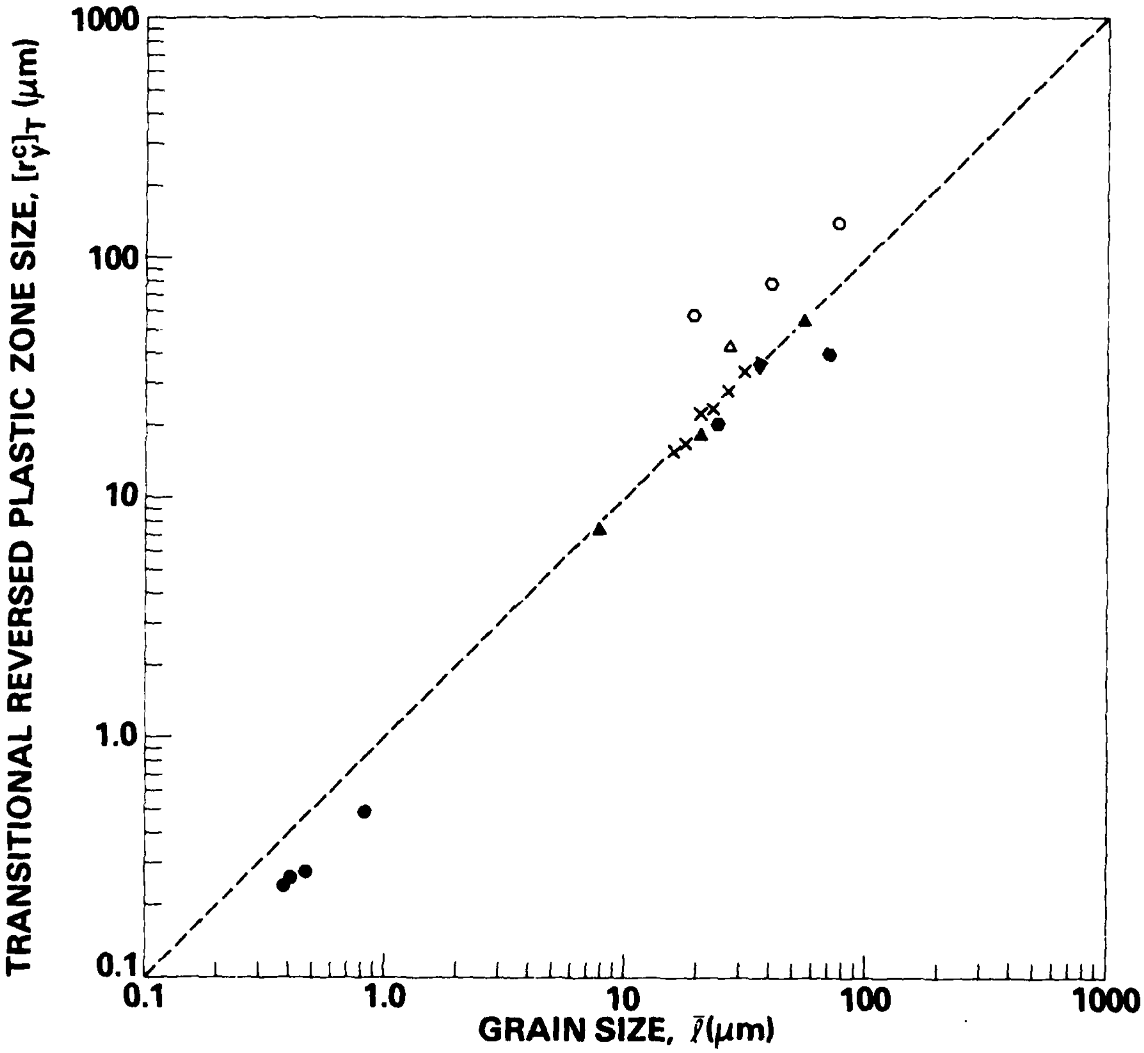

Fig. 8 Comparison of cyclic (reversed) plastic zone size at the transition point in bilinear growth-rate curve, $[r_y^c]_T$, vs. effective grain size ($\bar{l}$).

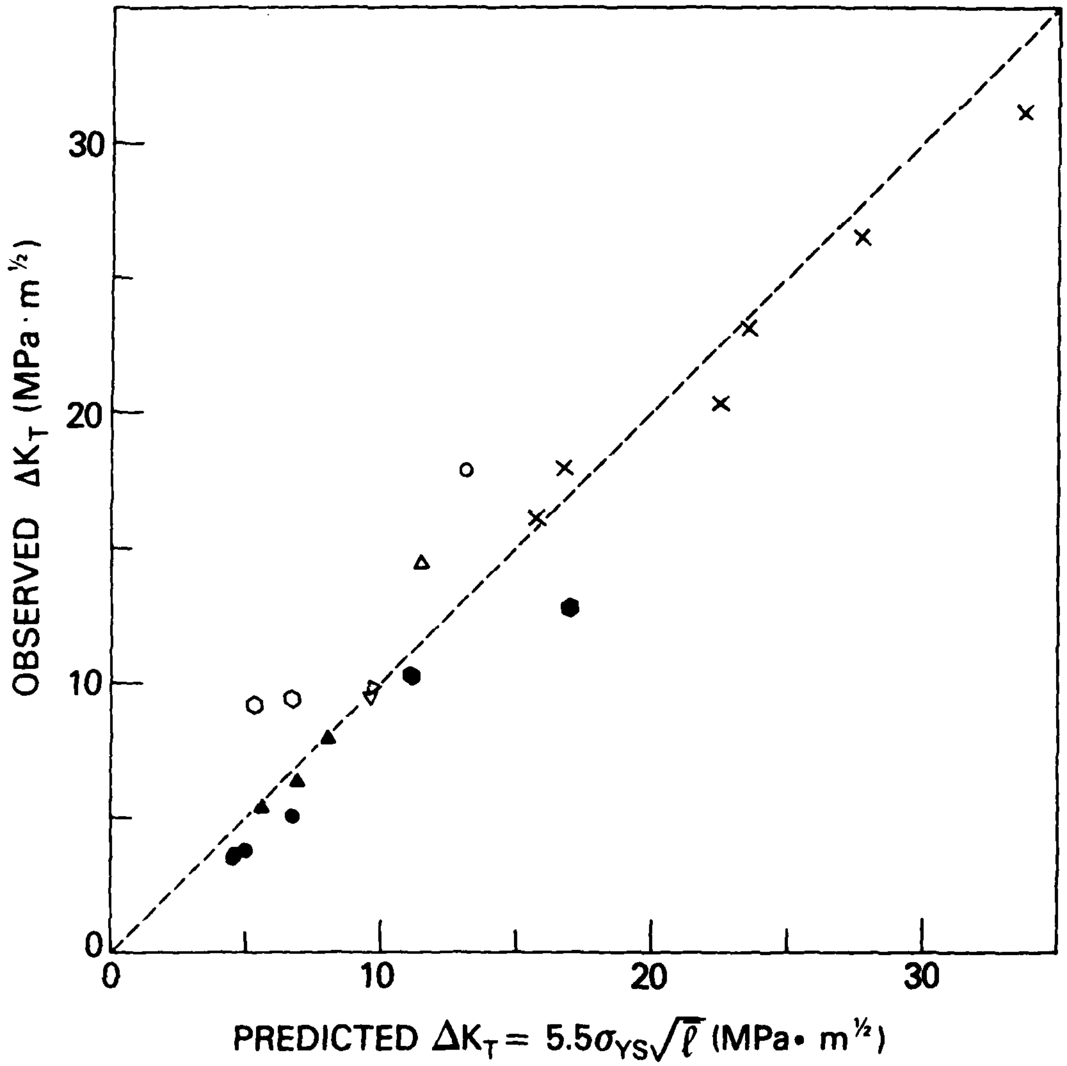

Fig. 9 Comparison of observed values of transitional stress-intensity range (ΔK_T) vs. predicted values from model according to Eq. (3).